

REMARKS

Applicants would like to thank the Examiner for extending the courtesy of an interview on July 2, 2007. Applicants' representatives, Dr. Gorte and Dr. Paz discussed the prior art and possible amendments to the claims.

Claims 1-30 and 54-61 are currently pending. Claims 31-53 were previously cancelled. Applicants reserve the right to pursue cancelled subject matter in one or more divisional applications. Claims 1, 20, 54 and 57-61 are currently amended. Support for the amendments may be found throughout the specification as filed, for instance at page 11, line 4 to page 12, line 6.

Rejections Under 35 U.S.C. §102(b)

Applicants appreciate with thanks the Examiner's withdrawal of the previous rejections under 35 U.S.C. §102(b).

Claims 57-61 are rejected under 35 U.S.C. §102(b) as being anticipated by Cable *et al.* (US Patent No. 5,589,285). The Examiner contends that Cable *et al.* teaches a solid oxide fuel cell comprising an anode, a cathode and a solid oxide electrolyte. The porous anode comprises a sulfur tolerant material (such as ceria) and optionally contains an electronically conducting phase, including copper. The Examiner further states that Cable *et al.* discloses a sulfur content in the hydrocarbon fuel of 1000 ppm or higher. The Examiner then concludes that Cable *et al.* anticipates Applicants' invention. Office Action page 2.

Applicants respectfully traverse this rejection for the following reasons. Cable *et al.* discloses a solid oxide fuel cell comprising an anode, a cathode and a solid oxide electrolyte. Contrary to the present invention, Cable *et al.* specifically discloses an anode material utilizing nickel. The inventors discovered that anodes containing at least copper and/or ceria are capable of directly processing sulfur containing fuels. The inventors discovered a method of incorporating relatively low melting materials (like copper) in an anode by initially co-firing the anode layer and the electrolyte layer, impregnating the resulting anode layer with a solution containing a salt of the anode material, and then heating the impregnated anode to drive off the liquid. Conventional methods of preparing anodes containing nickel (for example, nickel cermet was combined with YSZ and co-fired together with the electrolyte layer at temperatures at or about 1250°C) were not suitable for preparing an anode containing copper since the copper

would melt and diffuse out of the ceramic material during sintering. The inventors discovered that that anode needed to be sintered first, and then impregnated with a solution containing a salt of the anode material (e.g., Cu or CeO_2). It was by this method that the inventors were able to prepare a solid oxide fuel cell capable of directly oxidizing sulfur-containing fuels. Preparation of an anode containing no nickel prevents carbon formation as discussed in the introduction section of the present specification. The nickel free anode also produces a sulfur tolerant fuel cell able to operate directly with sulfur-containing fuels.

The inventors have also found that ensuring good contact between the ion-conducting electrolyte and the anode is critical for providing a high performance fuel cell. The present invention provides a process which bonds the electrolyte and anode layers. In one embodiment, the unsintered mixture of electronically-conductive and ionically-conductive materials is deposited on a layer of ionically-conductive electrolyte materials prior to being sintered to ensure sufficient contact between the layers. The two layers are then heated to a temperature of 1500°C (i.e., sintered simultaneously). In a second embodiment, the mixture is deposited onto a previously sintered layer of electrolyte and then sintered. See Specification, lines 4-13. This process provides optimal contact between the anode and electrolyte layers.

The fuel cell of Cable *et al.* is not bonded. The performance of the Cable *et al.* fuel cell would be so poor it would be rendered non-operative. Applicants respectfully submit two references in support of this conclusion. Zhao *et al.* "Dependence of Polarization in Anode-Supported Solid Oxide Fuel Cells on Various Cell Parameters." *Journal of Power Sciences*, 141:79-95 (2005) explains that the interpenetration of the electrolyte and the electrode is crucial for good performance in solid oxide fuel cells (see page 84, right column, 8th line). The non-bonded electrode has no interconnecting regions between the electrolyte and the electrodes. See also Lashtabeg *et al.* "Solid Oxide Fuel Cells-A Challenge for Materials Chemists?" *Journal of Materials Chemistry* 16:3161-3170 (2006).

Applicants believe that the invention as presently claimed is novel over Cable *et al.* However, solely in the interest of advancing prosecution, independent claims 57, 59 and 61 have been amended to recite "a porous anode comprised of a porous material containing no nickel". In addition, the claims have been amended to recite "wherein like particles of the anode and the solid electrolyte are bonded together". The solid oxide fuel cell of claims 57, 59 and 61 is

therefore novel over Cable *et al.* for the reasons articulated above. Withdrawal of the rejection is respectfully requested.

Rejections Under 35 U.S.C. §103(a)

Applicants appreciate with thanks the Examiner's withdrawal of the previous rejections under 35 U.S.C. §103(a).

Claims 1, 15-20, 30, 57, and 59 are rejected under 35 U.S.C. §103(a) as being unpatentable over Wallin (US Patent No. 6,017,647) in view of Isenberg (US Patent No. 4,812,329). The Examiner contends that Wallin teaches a solid oxide fuel cell comprising an electrolyte membrane of a ceramic oxygen ion conductor, a porous anode in contact with the electrolyte membrane on the fuel side and a porous cathode in contact with the electrolyte membrane on the oxidant side of the cell. The Examiner then states that the anode is typically a ceramic-metal composite. The Examiner concedes that Wallin does not teach the deposition of ceria in the porous anode structure. However, the Examiner claims that Isenberg teaches a method of making a sulfur tolerant composite cermet electrode for a solid oxide fuels cell. The Examiner states that this porous anode is impregnated with a solution of cerium nitrate and lanthanum nitrate followed by heat treatment to produce the ceria. The Examiner then concludes that it would have been obvious to one of ordinary skill in the art to include ceria deposited in the porous anode structure of Wallin because Isenberg teaches the resulting coated electrode has better sulfur tolerance. In addition, the Examiner states that the fuel cell of Wallin/Isenberg could be used in conjunction with a fuel with a sulfur content from about 1 to about 5000 ppm because Cable *et al.* discloses that impregnation of the anode with cerium oxide can utilize a fuel with a sulfur content up to 50 ppm. Office Action pages 2-3.

In raising an obviousness rejection under 35 U.S.C. §103(a), the Examiner has the burden of establishing that at the time of the invention, there was: (1) some suggestion or motivation to combine the reference teachings; (2) a reasonable expectation of success; and (3) that the prior art references, when combined, teach or suggest all the claim limitations. See MPEP §2143 (Eighth Edition, incorporating Revision No. 5, August 2006). Obviousness may not be established based upon hindsight or the teachings or suggestions of the inventor. *W. L. Gore and Assocs., Inc. v. Garlock, Inc.*, 721 F.2d 1540, 1551, 1553 (Fed. Cir. 1983); *Ruiz v. A. B. Chance Co.*, 357 F.3d 1270, 1276 (Fed. Cir. 2004). Here, the Examiner has not met this burden. It is

respectfully submitted that the obviousness rejection fails on each of the three grounds, as detailed below.

Applicants respectfully disagree with the Examiner's contentions. As explained above, the anode of the present invention does not contain nickel. In contrast (and as noted by the Examiner during the interview of July 2, 2007) the fuel cell of Wallin specifically utilizes an anode comprising nickel. Isenberg does not cure the deficiencies of Wallin. The anode described in Isenberg is a traditional nickel anode coated with a ceramic oxide. The coating is added in an attempt to protect the nickel anode from carbon formation. The coating is insufficient to fully protect the nickel anode at the operating temperatures contemplated by Applicants present invention (at least 700°C). Carbon formation would then take place while utilizing untreated sulfur containing fuels. One would not be motivated to combine the fuel cells of Wallin and Isenberg because the resulting anode would not provide sulfur tolerance. There would be no expectation that the fuel cell would operated successfully with untreated sulfur containing fuels, as claimed in Applicants' present invention. Applicants respectfully request withdrawal of the rejection.

Claims 2-14 and 21-29 are rejected under 35 U.S.C. §103(a) as being unpatentable over Wallin, Isenberg and Cable et al as applied above and further in view of Anumakonda *et al.* (US Patent No. 6,221,280). The Examiner concedes that the fuel cell of Wallin/Isenberg, as described above, does not teach the use of sulfur containing fuels. The Examiner contends that Anumakonda *et al.* teaches the use of sulfur-containing hydrocarbon fuels for a solid oxide fuel cell. The Examiner then concludes that it would have been obvious to one of ordinary skill in the art to utilize a fuel having a sulfur content of at least 50 to 3000 ppm with the fuel cell of Wallin/Isenberg because Anumakonda *et al.* teach the conversion of commercially available heavy hydrocarbon fuels to hydrogen. Office Action page 4-5.

The Examiner continues by stating that it is well known in the fuel cell art that methane (CH₄) and alcohols, such as methanol and ethanol, are functionally equivalent hydrocarbon fuels. Citing Patel (US Patent No. 4,791,033) and Keegan (US Patent No. 6,423,896), the Examiner concludes that it would have been obvious to one of ordinary skill in the art to substitute methanol for methane as the fuel in the operation of the solid oxide fuel cell disclosed by Wallin/Isenberg. Office Action page 5.

Applicants respectfully traverse the rejection. Applicants previously explained why Cable *et al.* is not an anticipating reference because Cable *et al.* comprises a nickel containing anode. The combination of Wallin and Isenberg does not add to Cable *et al.* and does not result in Applicants invention as articulated above. The Wallin, Isenberg and Cable *et al.* combination does not result in a solid oxide fuel cell that directly operates with a sulfur-containing hydrocarbon fuel that does not have to undergo prior treatment to remove organic sulfur compounds comprising a porous anode containing no nickel.

Anumakonda *et al.* does not cure the deficiencies of the combination. Applicants' invention comprises a fuel cell that is capable of processing sulfur-containing hydrocarbon fuels without the need to reform the fuel prior to introduction into the fuel cell. As stated in the Background of the Invention section of the specification, it has been proposed to replace hydrogen with commercially available and more economical hydrocarbon fuels, but that such raw fuels "are not currently in use as a fuel source suitable for a fuel cell because these fuel cells contain relatively high levels of sulfur" (page 2, lines 19-23). Anumakonda *et al.* describes a mechanism of reforming sulfur-containing fuels into hydrogen gas and carbon monoxide, *prior* to feeding the hydrogen gas to a solid oxide fuel cell.

The fuel cell of the present invention is capable of processing a sulfur-containing fuel without the need for prior reformation, as disclosed in Anumakonda *et al.* Applicants therefore claim a solid oxide fuel cell that includes a fuel that includes a sulfur-containing hydrocarbon having a sulfur content of from about 1 ppm to about 5000 ppm. Anumakonda *et al.* neither discloses or suggests a solid oxide fuel cell capable of directly processing, without reformation prior to introduction into the fuel cell, a sulfur-containing hydrocarbon as recited in the present claims. The Anumakonda *et al.* device is a sulfur removal process, not a simplified fuel cell. A person of skill in the art would not be motivated to modify the Wallin, Isenberg and Cable *et al.* device by including a two unit operation (vaporizer/mixer 21 and reactor 13) prior to the fuel cell. Combining the four references complicates the system. The combination of Wallin, Isenberg, Cable *et al.* and Anumakonda *et al.* increases the complexity of the system and does not result in Applicants' claimed invention of a solid oxide fuel cell that directly operates with a sulfur-containing hydrocarbon fuel that does not have to undergo prior treatment to remove organic sulfur compounds.

Applicants respectfully disagree with the Examiner's contention that methane (CH₄) and alcohols, such as methanol and ethanol, are functionally equivalent hydrocarbon fuels. Applicants submit concurrently with this Response and Amendment two articles describing the differences between hydrocarbon and hydrogen fuels. Sasaki *et al.*, "Direct-Alcohol SOFCs: Current-Voltage Characteristics and Fuel Gas Compositions." *Journal of the Electrochemical Society*, 151(7): A965-A970 (2004) explains that one would expect the hydrocarbon fuels to react differently than other fuels when used in solid oxide fuel cells. *See also* Gupta *et al.*, "Comparison of Conversion and Deposit Formation of Ethanol and Butane Under SOFC Conditions." *Journal of Power Sciences* 158:497-503 (2006). The paper by Gupta, *et al.* demonstrates that there are large differences between the various hydrocarbons and alcohols' in their tendencies to form carbonaceous deposits that deactivate the fuel cells.

Applicants also disagree with the Examiner's conclusion that the Patel and Keegan patents support the conclusion that it would have been obvious to one of ordinary skill in the art to substitute methanol for methane as the fuel in the operation of the solid oxide fuel cell disclosed by Wallin/Isenberg. Applicants agree that a hydrocarbon may be reformed. However, the fuel cell of Applicants' present invention does not require the reforming of fuel prior to introduction into the cell. As described in detail in Applicants' specification, carbon formation on nickel is possible even with steam reforming catalysts (which operate at a temperature similar to that used in solid oxide fuel cells). In addition, the molecular weight and steam:carbon ratios are important factors for consideration when determining the operation conditions necessary for steam reforming. *See, for example*, Armor, J. N. "Review: The Multiple Roles for Catalysts in the Production of H₂." *Applied Catalysts A: General* 176:159-176(1999). Here, the authors discuss how steam reforming is carried out commercially with methane as the fuel. It is noteworthy that the conditions required for steam reforming of methane involve temperatures that are typically greater than 800°C. This is in sharp contrast to the a temperature range of 160 to 310°C that is reported for steam reforming of methanol (C. Cao, G. Xia, J. Holladay, E. Jones, and Y. Wang, *Appl. Catal. A* 262 (2004) 19, explaining that not all hydrocarbons react in a similar manner while undergoing steam reformation). Finally, Ming *et al.* "Steam Reforming of Hydrocarbon Fuels." *Catalysis Today* 77:51-64 (2002) indicates that steam reforming of liquid hydrocarbons is much more difficult than steam reforming of methane.

For at least the reasons articulated above, Applicants respectfully request withdrawal of the rejection.

Claim 54 is rejected under 35 U.S.C. §103(a) as being unpatentable over Wallin in view of Cable *et al.* The Examiner characterizes Wallin as above. The Examiner concedes that Wallin does not disclose the deposition of copper in the porous anode structure. The Examiner claims that Cable *et al.* teaches a fuel cell wherein the porous anode comprises sulfur tolerant material (such as ceria) optionally containing an electronically conducting phase including copper. The Examiner then concludes that it would have been obvious to one of ordinary skill in the art to deposit ceria and copper in the anode of Wallin because Cable *et al.* teach the resulting anode will have better sulfur tolerance. Office Action page 6.

Applicants respectfully traverse the rejection. As discussed previously, the anode of the present invention does not contain nickel. Claim 54 has been amended to recite “a porous ceramic-metal composite anode containing (i) no nickel”. In contrast and as noted by the Examiner during the interview of July 2, 2007, the fuel cells of both Wallin and Cable *et al.* both utilize an anode comprising nickel. It is clear that the combination of Wallin and Cable *et al.* does not result in Applicants’ invention as articulated in claim 54, specifically, a solid oxide fuel cell that directly operates with a sulfur-containing hydrocarbon fuel that does not have to undergo prior treatment to remove organic sulfur compounds comprising a porous ceramic-metal composite anode containing no nickel. There is no motivation to combine the references of Wallin and Cable *et al.* because, as described in detail above, the resulting anode would contain nickel and where therefore not be sulfur tolerant. Withdrawal of the rejection is respectfully requested.

Claims 55, 56, 58 and 60 are rejected under 35 U.S.C. §103(a) as being unpatentable over Wallin and Isenberg as applied above to claims 1, 15-20, 30, 57 and 59 and further in view of Cable *et al.* The Examiner contends that Wallin and Isenberg disclose a fuel cell as described above. The Examiner concedes that Wallin and Isenberg do not teach or suggest a solid oxide fuel cell comprising a porous anode structure further comprising copper. The Examiner then states that Cable *et al.* teach a porous anode comprising a sulfur tolerant material, such as ceria, which optionally contains copper. The Examiner concludes that it would have been obvious to one of ordinary skill in the art to deposit ceria and copper in the porous structure of Wallin/Isenberg because Cable *et al.* teach that the resulting anode has better sulfur tolerance.

The rejection is respectfully traversed. Claims 55, 56, 58 and 60 depend from independent claims 1, 20, 57 and 59. The dependent claims include all the limitation of the independent claims from which they depend. As discussed previously, Claims 1, 20, 57 and 59 have been amended to include a porous ceramic metal composite anode comprising no nickel. The fuel cell of Wallin specifically utilizes an anode comprising nickel. The anode described in Isenberg is a traditional nickel anode coated with a ceramic oxide. Cable *et al.* does not cure the deficiencies of the Wallin/Isenberg combination because Cable *et al.* specifically discloses an anode material utilizing nickel. Carbon formation would then take place on the sulfur containing anodes while utilizing untreated sulfur containing fuels. One would not be motivated to combine the fuel cells of Wallin, Isenberg and Cable *et al.* because the resulting anode would not provide sulfur tolerance. There would be no expectation that the fuel cell would operate successfully with untreated sulfur containing fuels, as claimed in Applicants' present invention. Applicants respectfully request withdrawal of the rejection.

CONCLUSION

A petition for a three-month extension of time and the required fee are being transmitted concurrently with this submission. In the event that additional fees are deemed necessary, or in the event of any variance between the amount enclosed and the fees determined by the USPTO, please charge or credit any such variance to the undersigned's Deposit Account No. 50-0206.

In view of at least the foregoing, Applicants respectfully submit that the claims are in condition for allowance. Early notification of a favorable consideration is respectfully requested. In the event any issues remain, Applicants would appreciate the courtesy of a telephone call to their counsel at the number listed below to resolve such issues and place all claims in condition for allowance.

Respectfully submitted,

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